



Optimization of the Air Cleaning Properties of Fog

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Abstract

Fog droplets are very often used as a cleaning agent when air pollution can be dangerous for health conditions and ecosystem. This work presents a new system to optimize the cleaning properties of fog by tuning its microphysical parameters. For this purpose, a newly developed system, which is based on the electromagnetic echo effect (EMEE) sensor, is used to detect the most efficient interaction between fog and impurities, i.e., which fog droplets can be used to most effectively clean a certain type of pollutant from the air. Fog droplet spectra controlled by the nozzle pressure system can be used to effectively remove pollutants from the air. For this purpose, an automated system for aerosol generation can allow an accurate control over the fog microphysical parameters and the use of fluids with specific concentrations of pulverized chemical compounds. Fog droplet size distribution is controlled by the feeding gas pressure at the nozzle and chemical simulants. The experimental results showed that the microphysical parameters (MP) are directly related to the impurity of species used in the cleanup simulation process. The MP parameters of fog are liquid water content (LWC), droplet mean radius (R_m), droplet number concentration (N_d), and both aerosol type and mass concentration. In the lab testing, harmless simulants of CBRN (chemical, biological, radiological and nuclear) species were used. During the tests, fog droplet size distribution is controlled by the air pressure at the nozzle and simulants. It is concluded that an integrated fog generator system (IFGS) with EMEE sensor developed in the current work can be utilized broadly to control fog microphysical parameters, leading to an optimum aerosol/chemical species' cleaning process.

Keywords Pollution settling and removal · Electromagnetic echo effect · Chemical composition and impurities · Fog droplet spectra

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1 Introduction

Air pollution is a serious global problem (Brook et al. 2003) and adversely affects human health. Aerosol removal from the atmospheric boundary layer can be very important for cleaning the air. Aerosols can also directly and indirectly affect our climate (McFarquhar et al. 2011; Penner et al. 2001) by changing cloud microphysical properties (Tao et al. 2012; Gultepe et al. 2017b, a).

Artificially generated aerosols can be dangerous for human health that is a part of the ecosystem. The lethal power of polluting agents and their ability to cause mass fatalities rely on their dispersion and diffusion in a complex weather environment. Aerosols are those agents dispersed in air that can penetrate into the lungs. For example, casualties caused by inhaling Anthrax spores dispersed in air are two orders of magnitude greater than those caused by a cutaneous infection of the same spores (Malizia and

D'arienzo 2018). Pollution of the aerosols such as chemical and radiation agents can also be critical for human health (Goel 2015).

The impact of a dispersed chemical element on health can be reduced using various levels of fog physical conditions. This is usually true due to the exponential increase of the surface-to-volume ratio for decreasing sizes of fog droplets (Gultepe and Isaac 1996; Gultepe et al. 2006). The use of fine-dispersed emulsions (Brook et al. 2003) and small fog droplets (Tsujita et al. 2003) has been proven highly efficient and environmentally friendly for decontaminating equipment and complex facilities. The fog usage for cleaning poisonous aerosols from the air minimizes the amount of decontaminant required for cleaning procedures. Also, fog can be compatible with electrical and electronic equipment without any damages. Further, fog droplets can penetrate into complex holes and ducts, as well as venting ducts; therefore, it can be very useful for microclimate applications to remove harmful aerosols and bacteria.

Interestingly, Lee et al. demonstrate that hydrogen peroxide (H_2O_2) is spontaneously produced in micrometer-sized water droplets, generated by atomizing bulk water using nebulization, without the necessity of any chemical reagent, catalyst, or applied electric potential (Lee et al. 2019a, b). H_2O_2 production increases with decreasing microdroplet size. It is later reported (Lee et al., 2020), that the H_2O_2 can also be spontaneously generated in water microdroplets formed by condensation of water vapor from air on a cold surface. In addition, Lin and co-authors (Lin et al. 2023) uncover a size-dependent nature of the charge transfer between water microdroplets. The results could be used for green and inexpensive production of H_2O_2 for safe air pollution control (see, e.g., Deo 1988).

It is well known that both fog and rain, because of their washout effects, can be used as air cleaning agents, since they both have higher fall velocities compared to pollutants (Pilat et al. 1974; Guo et al. 2016). Recently, a study by Ardon-Dryer et al. (2015) suggested that rain having with large drops (> 100 micron) can be very effective for cleaning air in the atmosphere. It was discovered that, in general, the smaller the size of falling rain drops in the same mass concentration as in large drops, attracts many more aerosol particles, especially at lower relative humidity (RH) conditions. Moreover, a device designed for fast and massive decontamination of aerosols through the generation of sub-micron fog jets has been developed and tested successfully for removing the CBRN clouds (Pérez-Díaz et al. 2018). A precise assessment of the “cleaning” process requires a precise characterization of the fog microphysical properties.

Using various air pressures at the nozzle in the climatic wind tunnels (Gultepe et al. 2019; Qing et al. 2018), aerosol number concentration (N_d) and mean diameter (D_m) of the droplet size distribution (DSD) are obtained when

pulverizing fluids with specific mixture of the mass concentrations of chemical compounds. The sensors used for this purpose in the past have been operated on the basis of the “laser-induced charge effect” (Ivanov and Konstantinov 2002; Ivanov and Kuneva 2011). The study of Ivanov et al. (2020a) suggested that the name of this laser-induced charge effect should be changed to electromagnetic echo effect (EMEE) because this reflects better the nature of the effect. In this way, it cannot be confused with the photoelectric effect used commonly. The EMEE is represented by small, alternating voltage, and detected using targeted EMF on all kinds of substances (Ivanov et al. 1995; Park et al. 1996). The same technique has been also used for numerous applications (Ivanov and Kuneva 2011; Abbate et al. 1995; Ivanov and Konstantinov 1999; Ivanov 2004; Pérez-Díaz and Kuneva 2017) and also used for the development of fog sensors used for industrial applications. The new methods for contactless evaluation of fog microphysical parameters in artificially produced sprays have been previously used to detect aerosol impurities in the composition of fog (Ivanov et al. 2017a, b, 2020a, b, 2021; Ivanov and Pérez-Díaz 2020).

The analyzing of the fog-chemical composition in the atmosphere has been carried out by collecting the droplets from special mesh collectors and testing them in a laboratory, which is difficult and time consuming (Klemm et al. 2012). Earlier studies mainly focused on the chemical properties of the air pollution aerosols and their nucleation capability used in numerical studies (Tsujita et al. 2003; Marquez-Viloria et al. 2016) but not in the fog-aerosol interactions. Lately, time of flight fog-aerosol analysis is possible and used in the cloud droplet research (Hwajin et al. 2019).

The current study is related to the COUNTERFOG project which is funded by the European Commission (PN 312804) that aims to create a system for large-scale decontamination of air using fog droplets. The chemical pollutants are chosen as harmless simulants of the main types of CBRN agents. Since it was not possible for us to measure in real time the actual aerosol removal from air by using fogs (this requires very expensive equipment such as fog/cloud chambers), we conducted the experiments indirectly—we used the EMEE sensor system that is designed to measure in real time the relative amount of pollutants in the composition of various fogs. In this way, we can determine the parameters of fog droplets that have absorbed optimal quantities of pollutants, and these results can be used in real-life applications regarding aerosol removal from air. The fog generating systems can be designed and tuned to produce fog droplets with the required parameters for optimal removal of certain pollutants from the air.

The paper is structured as follows: Sect. 2 represents the experimental setup and an integrated fog generator system

(IFGS). Sections 3 and 4 focus on the results and discussions. Section 5 provides the main conclusions.

2 System Setup and Measurements

The IFGS generates a controlled fog droplet size spectrum in a specific sampling volume. Artificial fog can be used as a cleaning agent to remove aerosols from the sampled air. To accomplish this goal, the purposed platform uses the EMEE sensor that measures the relative amount of pollution aerosols collected by the fog.

The relationship between fog physical parameters and aerosol chemical composition is investigated using the IFGS that operates on the basis of the EMEE. This IFGS prototype system, using various thresholds for chemical species, is able to generate aerosols with different chemical compositions that can lead to dedicated droplet size (R_m) and number concentration (N_d). The IFGS is designed to work with up to six different fluids and durations of spraying from 0.2 to 5.5 s. An important advantage of this system is that it provides a fast and easy way to pulverize different chemical solutions, and offers a precise control of the amounts of impurities in the fluids. As a result, generated fog can have various particle size distributions (PSDs) with changing LWC and effective size. The mixing of chemical species with pure water is not dispersed in air but are dissolved in the pure liquid water before releasing into the air.

Figure 1 presents the automated IFGS. This system includes following units: the fog generator, fog measuring equipment, spray control unit with a timer, and signal detector unit. The flow diagram for the IFGS is shown in Fig. 2.

The numbers in Fig. 2 indicate the location of the ten units in the IFGS. The unit 1 is the receiver of the optical sensor used for fog droplet spectra measurements. The unit 2

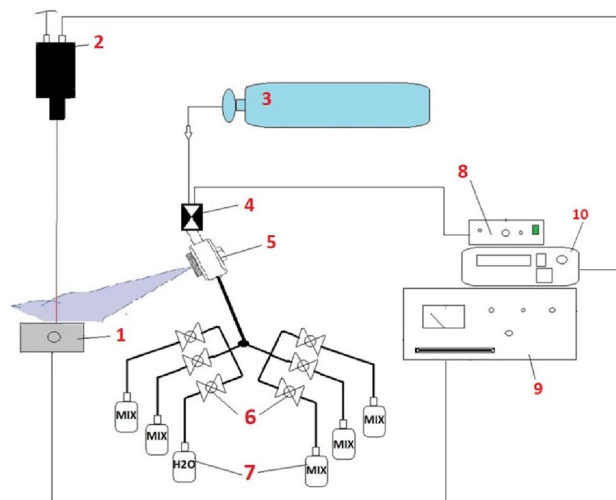
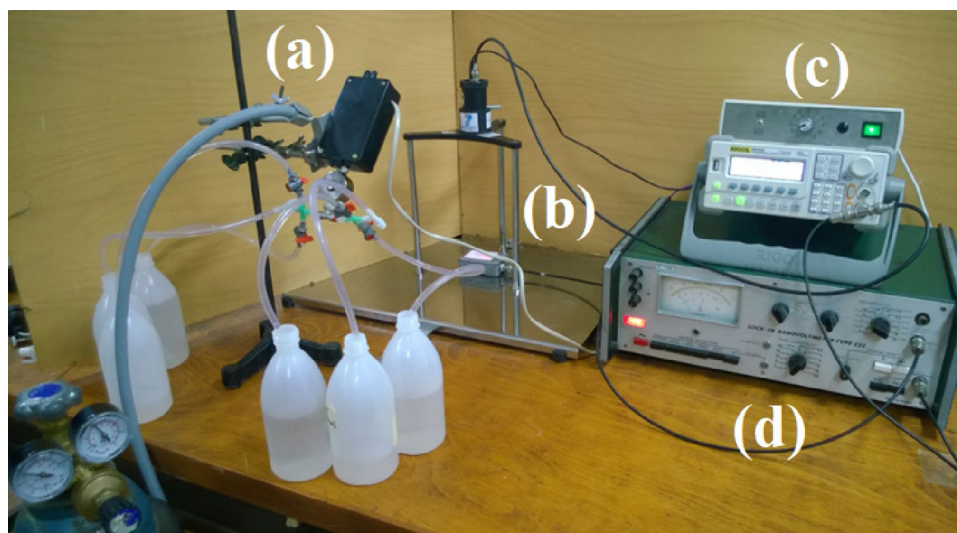


Fig. 2 Block diagram of the automated system for generation of fog with impurities and the fog sensor – (1) laser sensor transmitter; (2) laser sensor receiver unit; (3) gas pressure tank; (4) valve; (5) atomizing nozzle; (6) valves; (7) fluid containers; (8) spraying control unit; (9) lock-in nanovoltmeter; (10) frequency generator

is for the optical sensor transmitter. The unit 3 is for the generation of the gas (e.g., nitrogen) used under pressure that is fed from the tank (3) to the valve (unit 4), and then lead to the atomizing nozzle (Unit 5). The prototype IFGS has six valves (unit 6), so that switching between different containers representing various chemical species can be done quickly and easily. Thus, pulling water from a selected container (7) can be mixed with selected chemical species. The diameter of the nozzle is 0.4 mm with a spraying angle of 20° (suggested by the manufacturer, Lechler GmbH, Metzingen, Germany, <https://www.lechler.com/de-en/>). The nozzle (model #136.316.35.A2) is a pneumatic brass-plated atomizing nozzle, which works on the siphon/gravity principle.

Fig. 1 Integrated fog generation system (IFGS): **a** fog generation, **b** fog measuring unit, **c** spraying control unit, and **d** signal detector unit



A manometer and valves were mounted on the pressure gas tank, which allowed to precisely control the feeding pressure of the nitrogen gas, so that different characteristics of the generated fog can be obtained, such as the number concentration (N_d) and droplet size (R_m). Depending on the impurity (defined as a chemical composition) of a selected species, a specific container is selected to generate a specific fog type. The electromagnetic valve (unit 4) is controlled by a pulse generator (unit 8) and was designed locally. Different settings by unit 8 can be used to achieve desired durations of fog spraying. Fog produced by the nozzle is directed into the sensor sampling area consisting of a laser (unit 2) and a frequency generator (unit 10). The unit 10 modulates the transmitted laser intensity and its received absorption after fog extinction. The signal from the sensor is measured by a lock-in nanovoltmeter (9).

3 Method

In the analysis, a certain amount of pure water is mixed with a chemical species as well as nitrogen gas in the unit 5 nozzle system. The control unit generates uniform impulses at either 0.2- or 0.5-s increments from 0.2 to 5.5 s. When the desired values for the impulse duration are set, the generation of an impulse with the desired time duration is initiated via a button. This impulse is transferred to the electromagnetic valve (unit 4), and thus a fog spraying through the nozzle is generated.

For the experiments, four kinds of chemical species were dissolved in distilled water—potassium dihydrogen phosphate (KH_2PO_4), urea ($\text{CO}(\text{NH}_2)_2$), potassium hexacyanoferrate trihydrate ($\text{K}_3[\text{Fe}(\text{CN})_6]$), and Cefazolin (an antibiotic with symbol of $\text{C}_{14}\text{H}_{14}\text{N}_8\text{O}_4\text{S}_3$). The chemical species were chosen, because they have a similar chemical structure as the CBRN (chemical, biological, radiological and nuclear) agents, but they are harmless to be used in a laboratory. This way, the presence of a pollutant in fog droplets can be simulated. For example, KH_2PO_4 has been selected as a harmless simulant of the CBRN chemical poisonous agents. In a water solution, it dissociates to give phosphate ions which resemble a part of the abovementioned agents' molecules.

$\text{K}_3[\text{Fe}(\text{CN})_6]$ was chosen as a chemical agent simulant due to the presence of cyanide ions in its structure. They are bonded in a stable complex with iron, which makes the compound safe to work with (its solution is also used in photography). The cyanide functional group is a constituent of the blood agents Cyanogen Chloride and Hydrogen Cyanide.

The prepared solutions were used to generate fog DSD with different droplet number concentration N_d and mean diameter (D_m) at 2, 3 and 4 bar of feeding gas pressure (P). The N_d , liquid water content LWC, and droplet mean radius R_m are obtained from DSD assuming a relationship between nozzle pressure at the liquid exit and droplet characteristic size. The solution characteristics at each gas pressure were measured using a laser particle size analyzer (LPSA uses a 0.532-micron wavelength and it is the Winner 319A model, manufactured by Winner Particle Instrument Stock Co. Ltd., Jinan, China, <http://en.jnwinner.com/>) at a distance of 1 m. The LPSA has the measurement size range of 1–500 micron at 50 channels. For each case, 100 of the measurements are taken at 1 Hz sampling rate and then averaged. The duration (dt) of each spraying is selected as 0.4 s. For each gas pressure, six measurements of the signal intensity were recorded and then its average value for the dt is calculated. In the analysis, the droplet size of each solution (mean diameter, D_m) is obtained as a function of 3 pressure levels (e.g., 2, 3, and 4 bars, see Table 1). This procedure is repeated for each of the four solutions and results are summarized in next section.

4 Results

4.1 Solution Microphysical Characteristics

The results are summarized in Table 1 where D_m of DSD is measured by the particle size analyzer against the nozzle pressure (P) (from 2 to 4 bar) for each chemical species (four types and distilled water).

Table 1 shows that D_m decreases with gas pressure increasing. Larger D_m values are seen for the distilled water. On the other hand, cefazolin solution has a lower fog droplet D_m . It is clearly seen that D_m is significantly lower for

Table 1 Mean droplet diameter D_m versus nozzle pressure P for the 4 chemical solutions

	P (bar)	2	3	4
D_m of droplets (μm) of the solution containing:	Potassium hexacyanoferrate (III) trihydrate	13.7	8.4	8.0
	Urea (carbamide)	13.7	10.9	8.6
	Potassium dihydrogen phosphate	17.2	14.0	9.0
	Cefazolin	9.9	8.0	7.6
	Distilled water	26.6	22.4	17.6

mixtures compared to distilled water, indicating CCN effect on droplet formation.

The D_m versus nozzle pressure for the 4 chemical species are shown in Fig. 3. The fog mean diameters for each solution are obtained based on the three nozzle pressure levels. Results showed that increasing nozzle pressure leads to decreasing D_m which is consistent with theory.

It is clear to see that the presence of chemical species can alter the fog DSD, affecting the mean diameter. The fog generating system can be used to control fog microphysical properties including D_m , LWC, and N_d . Using the mass continuity concept of the fluid flow, the ratio of P_1 to P_2 (pressures in two different conditions) is assumed to be related to the ratio of N_{d1} to N_{d2} (droplet number concentration at P_1 and P_2). N_{d1} is assumed as a fixed value ($\sim 100 \text{ cm}^{-3}$) for typical marine fog conditions (Gultepe et al. 2017b). Then, N_{d2} is calculated based on mass continuity concept for a given air flow. In reality, N_d is usually a function of aerosol number concentration, aerosol composition, and RH. Table 2 shows the fog droplet mean volume radius (R_m), LWC, visibility (Vis), as well as flow rate (Q_r) and droplet speed (V_{dn})

at the nozzle. The dependence of LWC on R_m and N_d can be given by (Gultepe and Isaac 2004) as:

$$\text{LWC} = \frac{4}{3} \pi R_m^3 N_d \rho_w \tag{1}$$

where ρ_w is the water density. Then, Vis related to LWC and N_d based on in-situ measurements using FM100 instrument is obtained (Gultepe et al. 2006) as:

$$\text{Vis} = \frac{1.002}{(\text{LWC} \times N_d)^{0.6473}} \tag{2}$$

This equation represents marine fog conditions and may be used for a general testing of measured DSD relation to chemical agents. Equation 2 is used for Vis calculation as function of LWC, N_d , and R_m for each chemical agent, and the results are summarized in Table 2.

The results from Table 2 can be summarized as follows:

- Droplet number concentration is found to be critical for cleaning up the chemicals from the polluted environment

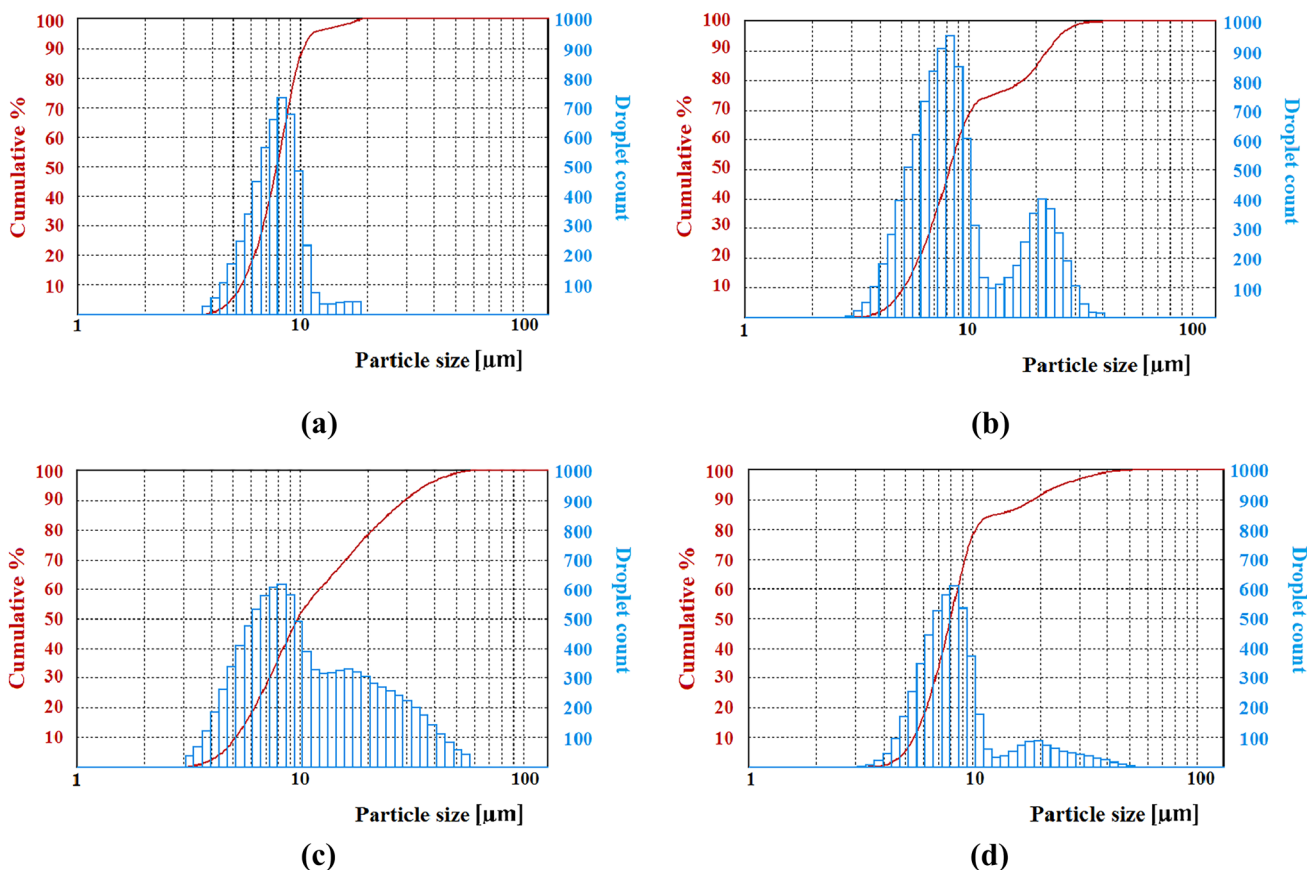


Fig. 3 Fog DSD with highest cleaning efficiency for pollutants (D_m versus droplet count): **a** potassium hexacyanoferrate trihydrate, **b** urea, **c** potassium dihydrogen phosphate, **d** Cefazolin

Table 2 EMEE signal deviation versus mean droplet radius (R_m) of fog DSD containing various chemical impurities at various gas pressures

Pulverizing gas pressure (bar)	2	3	4
Q_r Flow rate through the nozzle ($l\ h^{-1}$)	1.68	1.92	2.07
N_d ($\#\ cm^{-3}$)	122	139	150
V_{dn} Droplet speed through nozzle ($m\ s^{-1}$)	1.34	1.53	1.65
<i>5% potassium hexacyanoferrate trihydrate solution</i>			
R_m (μm)	6.9	4.2	4.0
EMEE signal deviation (%)	23.0	20.0	27.0
LWC ($g\ m^{-3}$)	0.167	0.043	0.040
Visibility (km)	0.14	0.31	0.31
<i>5% urea solution</i>			
R_m (μm)	6.9	5.5	4.3
EMEE signal deviation (%)	12.8	35.0	22.6
LWC ($g\ m^{-3}$)	0.167	0.097	0.050
Visibility (km)	0.14	0.19	0.27
<i>5% potassium dihydrogen phosphate solution</i>			
R_m (μm)	8.6	7.0	4.5
EMEE signal deviation (%)	12.0	143.7	60.0
LWC ($g\ m^{-3}$)	0.324	0.199	0.057
Visibility (km)	0.09	0.12	0.25
<i>1% Cefazolin</i>			
R_m (μm)	5.0	4.0	3.8
EMEE signal deviation (%)	113.3	86.5	19.8
LWC ($g\ m^{-3}$)	0.064	0.037	0.034
Visibility (km)	0.27	0.35	0.35
<i>Distilled water (for reference)</i>			
R_m (μm)	13.3	11.2	8.8
LWC ($g\ m^{-3}$)	1.198	0.815	0.427
Visibility (km)	0.04	0.05	0.07

because it is influenced by aerosol mass and size of the particles;

- Droplets with R_m of 4.0 μm can be ideal for cleaning potassium hexacyanoferrate from the polluted environment;
- R_m of 5.5 μm can be used to most effectively cleaning the urea;
- Potassium dihydrogen phosphate is most successfully collected by droplets with R_m of 7.0 μm ;
- The solution containing Cefazolin with R_m of 5.0 μm can be used most effectively clean the polluted air.

4.2 Solution Droplet Size Spectra

In this subsection, spectral characteristics of solution droplets measured by the laser particle size analyzer are studied based on the scatter plot of the particle size (D_m) versus cumulative count values and number counts (Fig. 3). Figure 3 presents measurements for each of the four types of

solutions. Based on the fog DSD in Fig. 3, the following points can be emphasized:

- The fog containing 40% of droplets with sizes between 3 and 10 μm , and 60% of droplets between 14 and 50 μm optimally collect potassium hexacyanoferrate;
- The fog that can be most effective in cleaning up urea from the air, contains about 75% of droplets between 3 and 13 μm , and 25% in the range 13–40 μm ;
- The fog that contains about 50% of its droplets having sizes between 3 and 10 μm , and the other 50%—between 10 and 55 μm , is most suitable for collecting potassium dihydrogen phosphate;
- The fog in which 80% of the droplets have diameters in the range 3.5–10 μm , and the other 20% are between 10 and 50 μm , can be very effective cleaning up Cefazolin from the air.

From the results presented above, it is clear that each type of pollutant can be cleared from air by fog with specific size distribution parameters, such as R_m , LWC, and N_d that represent a specific DSD.

5 Discussion

In this study, an automated aerosol-water droplet generation system is developed to test chemical impurities effect on fog droplets that is measured by an LPSA. The fog droplet spectra are obtained using four chemical species and a gas water mixture, and controlled by the nozzle system. Fog droplet generating solutions are initially made by dissolving the chemicals into distilled water (Fig. 2). Then, the nitrogen under pressure is connected to the pneumatic nozzle of the pulverizing system to draw the solution from the container to the nozzle, generating fog droplets. At first, we sprayed the mixture towards the EMEE sensor to get the signal deviation, and then the same fog is sprayed towards the LPSA to determine the particle size spectra. After that, we can compare the signal deviation with the respective mean diameter. The higher the deviation, the greater the amount of the impurity carried by the corresponding droplets. In that way we can determine the D_m with the highest amount of impurity.

In the experiment described above, major difficulties can be summarized as:

- Stability of the system with room RH and temperature T that were fixed at 85% and 25 $^{\circ}C$, respectively;
- If more than one chemical species is introduced into the composition of fog, the sensor at its current level of development would not be able to distinguish the concentration of each impurity separately. This will lead to large uncertainty in characteristic size of PSD;

- The most important issue is that the LPSA was not able to provide direct measurements of N_d and this led to analysis difficult to be applied, and in future, this will be improved with a new supporting particle sizer.

Despite these aforementioned issues, the presented system is capable of quick analysis of fog pollution concentrations, since the EMEE sensor operates in real time.

6 Conclusions

In this work, the IFGS is developed and operates on the basis of the EMEE signal recognition for the aim of measuring fog droplets properties and optimization of the pollution cleaning process. The IFGS is an automated system that is used for fog simulation experiments in the laboratory conditions. The experimental setup allows an efficient interaction between fog droplets and various assumed pollutant amounts. The four chemicals as shown in Table 1 are prepared to simulate hazardous pollutants effects on fog size distributions. The best fog-chemical species interactions are determined by detecting the highest EMEE signal deviations. These results show that specific fog parameters exist for each pollutant type and can be used to remove the pollutant species, in this case, CRBN chemicals species. Creating databases as discussed here can improve the capabilities for pollution eradication and retrieval procedures. The following conclusions from this work can be drawn:

- IFGS is found to be effectively used for fog generation with various PSD;
- Chemical species led to varying R_m and changing droplet number concentration;
- Droplet R_m was about 5–10 μm for urea and Cefazolin and 15–25 μm for potassium dihydrogen phosphate and potassium hexacyanoferrate trihydrate;
- Largest uncertainty in the analysis occurred when the environmental RH and T fluctuates about 20% of the fixed values.
- Results suggested that IFGS can be applicable for real environmental problems but needs to be further tested for varying environmental conditions.

It is suggested that creating proper databases related to fog formation as a function of various chemical species and controlled pressure systems can improve the capabilities for air pollution eradication and fog simulations. In future, as a continuation of the work within the COUNTERFOG project, we would like to simulate real atmospheric conditions to be tested for the removal process of dangerous chemical species.

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Declarations

Conflict of Interest The authors have no competing interests to declare that are relevant to the content of this article.

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